MASTER COPY KEEP THIS COPY FOR REPRODUCTION PURPOSES

AD-A249 798

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

(2)

...average 1 hour per response, including the time for reviewing instructions, searching existing data sources, ...the collection of information. Send comments regarding this burden estimate or any other aspect of this ...to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson ... Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (*Leave blank*)	2. REPORT DATE 1/10/92	3. REPORT TYPE AND DATES COVERED

4. TITLE AND SUBTITLE
"Research Studies on Extreme Ultraviolet and Soft X-ray Lasers"

5. FUNDING NUMBERS
ARO MIPR 107-91

6. AUTHOR(S)

S. E. Harris

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
ARO 26160.22-PH

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION/AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (*Maximum 200 words*)

This report describes studies concerned with the physics of short wavelength lasers and with applications of a new type of quantum mechanical interference. Highlights of this work include the operation of a traveling wave X-ray pump H_2 116 nm laser; the development of a new type of depletion spectroscopy for core-excited states; and the proposal of lasers without inversion and related concepts applicable to nonlinear optics.

14. SUBJECT TERMS			15. NUMBER OF PAGES 32
			16. PRICE CODE
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL

Research Studies on Extreme Ultraviolet and Soft X-ray Lasers

Final Report

S. E. Harris

25 October 1988 - 24 October 1991

U. S. Army Research Office

Contract No. F49620-88-C-0120

Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305

Table of Contents

Research Studies on Extreme Ultraviolet and Soft X-ray Lasers

Section 1

Introduction

This has been an unusually successful and diverse contract period. As the contract began, our efforts were focused on the invention and first demonstration of new types of lasers which operate in the vicinity of 100 nm. We were also concerned with basic spectroscopic studies of core-excited atoms which we hoped could be made to lase. Very early in the contract period, we were successful in the construction of a 12.8 eV laser which was pumped using the novel traveling wave geometry which had been invented under the earlier contract period. But we did not understand how this laser worked, and in attempting to understand it, we postulated the possibility of lasers which could operate without the need for a population inversion. It was this topic that formed the theme of much of the subsequent effort during the contract period. The laser without inversion concept was our first look at an interference concept which allowed ground-level atoms to be rendered transparent while at the same time retaining the emissive properties of excited atoms. A principal result was the realization that this type of non-reciprocal quantum mechanical effect could be obtained by applying an electromagnetic field to an atomic media. Toward the end of the contract period, a key experiment was performed: In this experiment, the transmission on an ultraviolet autoionizing transition in Sr was changed from exp(-20) to exp(-1). This transmission change was accomplished by applying a laser with a power density of about 10 MW/cm^2 between an otherwise empty level and the upper level of the transition which was to be rendered transparent. Of most importance, this transparency occurred with almost all of the population remaining in the ground level.

The type of transparency described above has possible application to lasers which may operate without population inversion and also to new types of nonlinear optical devices. The special feature of such nonlinear optical devices would be the possibility of greatly improved nonlinear coefficient times length products. There is also the possibility of creating new types of XUV radiation sources which have a brightness which is much higher than that of the effective temperature of the atom.

There was another substantial success as this contract period neared its close: we were able to operate an H_2 laser at 116 nm. This laser operated using our traveling wave geometry where an incident laser beam generated an intense traveling wave flux of X-rays, which in turn, by photoionization, created the electrons which pumped the H_2 laser. A feature of this laser was that the atoms remained at their ambient temperature in the presence of the hot electrons.

Section 2 of this report lists other inter-related contracts. Section 3 summarizes the principal contributions of this program. Section 4 describes our overall conclusions and gives some ideas of future work. Section 5 is a bibliography of the publications which have resulted from this contract. Section 6 gives the abstracts of these publications.

Section 2

Personnel and Relation to other Contracts

Professor James Young, who played a major role in this program, has left us to become a professor at Rice University. Dr. Guang-Yu Yin, who has been with us for five years, and is a superb laser engineer and experimentalist, has undertaken much of Professor Young's responsibility. Chris Barty has joined our group as an acting professor of Applied Physics.

The work described here has been jointly supported by the Air Force Office of Scientific Research, the U. S. Office of Naval Research, and by the Strategic Defense Initiative Organization. We have also obtained substantial help with equipment from Lawrence Livermore National Laboratory. The fsec laser system was built with Army-Air Force support.

Section 3

Summary of Contributions

The overall contributions of the program may be grouped into four categories:

XUV Laser Development

1. Experiments were performed which demonstrated that lasers with several joules of pumping energy could be used to generate sufficient X-rays to pump laboratory scale short wavelength lasers. Suggestions were made for Auger lasers and neutral species lasers. Following the first demonstration of the Xe Auger laser (109 nm) by Roger Falcone and his students at LLNL, Xe and Zn Auger lasers were demonstrated at several joules of pumping energy.

2. We invented and demonstrated a traveling-wave geometry which has been key to much of the recent success of this program. This geometry allows a long interaction length with matching of the group velocity of the pumping X-rays and the generated XUV laser beam, while maintaining high conversion of pump energy to incoherent soft X-rays.

3. Using this geometry, the fully saturated operation of the Xe Auger laser was shown.

4. The concept of pumping by X-ray produced electrons was suggested and demonstrated in Li.

5. The combination of the traveling-wave geometry and electron pumping was used to make the first laser whose upper level is above a lower continuum. This laser in Cs vapor operates at 96.9 nm and had an extrapolated small signal gain in excess of exp (80).

6. An improved version of the traveling-wave geometry was used to demonstrate lasing in molecular hydrogen at 116 nm. The Xe laser was operated at 2 Hz, and recently, operated by pumping with a modified commercial Nd:YAG laser (e.g. Quanta-Ray).

Conceptual and Spectroscopic

1. We suggested and demonstrated the technique of laser-depletion spectroscopy of core-excited levels. In this technique, a tunable laser is used to deplete the population of a radiating core-excited level, as other levels within the core-excited manifold are accessed. Working in neutral Rb, we measured level positions, linewidths, and autoionizing times, all to the highest accuracy ever obtained. This same work demonstrated levels with autoionizing times corresponding to linewidths which were well beneath the Doppler linewidths, and which had branching ratios for radiation as high as 70%.

2. The possibility of constructing lasers which could operate without the need for a population inversion was suggested. It was shown theoretically that two closely spaced lines could exhibit an interference in absorption that is absent in emission.

3. A method for, in effect, creating two closely spaced levels was proposed. In this method we use an electromagnetic field to couple a metastable level to an autoionizing or radiating level, and thereby dress the later level. We term this effect as electromagnetically induced transparency.

4. The possibility of greatly improved nonlinear optical processes, which operate by using this transparency to allow operation near what would otherwise be an opaque resonance was noted.

5. The generation of 104.8 nm radiation to a sharp absorption window in Zinc was demonstrated.

6. Very recently, working in neutral Sr, the opacity of an autoionizing transition was changed from exp(-20) to exp(-1), thus providing a first dramatic demonstration of a laser induced transparency.

Sub-Picosecond Laser System

An early version of a sub-picosecond laser system has been developed. The system consists of a synchronously pumped 85 fsec dye laser, followed by a Martinez type pulse expander, a Ti:Sapphire regenerative amplifier, and a grating compressor. We obtain about 60 mJ in 125 fsec in a beam that is 1.2 times - diffraction - limited. We estimate that, in the absence of other defocusing effects, this system will allow a focused power density of 10^{18} W/cm^2.

Generation of Incoherent X-rays

As this contract was ending we performed a striking set of experiments: By focusing a fsec laser onto a heavy metal target, we generated incoherent hard X-rays at an efficiency which approaches 10^{-3}. There appear to be tales of the X-ray radiation extending to beyond an MeV. Studies of efficiency and spectral and angular distribution are continuing.

Section 4

Conclusions and Future Direction

We believe that we have now carried our work on 100 nm lasers as far as makes sense at this time. We have tried unsuccessfully to take the Cs Auger laser to the several hundred angstrom region. The lasers that we have made are working very well and Professor James Young is continuing with their development and application in the Electrical Engineering department at Rice.

For the time being, we view our studies on depletion spectroscopy of the alkali atoms as complete.

We believe that the general area of quantum interference and its application to nonlinear optics and new types of laser devices is extremely promising. It is fortunate, so many years after the invention of the laser, to find this type of unexplored territory. Our basic approach will be to first attempt to delineate the basic principles using gas systems and then to seek application in solid state systems. Areas of application include lasers without inversion, nonlinear optical devices, and perhaps a study of some novel approaches to the manipulation of optical group velocities.

We will also aggressively continue our studies on the interaction of high-intensity fields with matter. We are interested in the use of high order harmonic generation, both in its own right, and to generate temporal structure on a scale of 10^{-17} sec.

Section 5

Bibliography

1. J. K. Spong, A. Imamoglu, R. Buffa, and S. E. Harris, "Laser-Depletion Spectroscopy of Core-Excited Levels of Neutral Rubidium," *Phys. Rev. A* **38**, 5618 (1988).

2. C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris, "12.8-eV Laser in Neutral Cesium," *Phys. Rev. Lett.* **61**, 2201 (1988).

3. S. E. Harris, "Non-Reciprocity of Autoionizing Interferences: Lasers Without Inversion," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2, pp. 414-417.

4. C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris, "12.8 eV Laser in Neutral Cesium," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applicatiors*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2. pp. 13-20.

5. J. D. Kmetec and S. E. Harris, "Targets for Efficient Femtosecond-Time-Scale X-Ray Generation," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2, pp. 209-212.

6. S. E. Harris and J. J. Macklin, "Lasers Without Inversion: Single-Atom Transient Response," *Phys. Rev. A*, **40**, 4135-4137 (October 1989).

7. S. E. Harris, A. Imamoglu, and J. J. Macklin, "Nonreciprocal Emissive and Absorptive Processes," in *Laser Spectroscopy IX*, M. S. Feld, J. E. Thomas, and A. Mooradian, eds. (Academic Press, Inc., San Diego, 1989), pp. 232-235.

8. A. Imamoglu and S. E. Harris, "Lasers Without Inversion: Interference of Dressed L.ifetime-Broadened States," *Opt. Lett.* **40**, 1344-1346 (December 1989).

9. J. K. Spong, A. Imamoglu, J. D. Kmetec, and S. E. Harris, "Extreme Ultraviolet Fluorescence from Core-Excited Levels of Neutral Rubidium," *Opt. Lett.* **14**, 168 (1989).

10. S. E. Harris, "Lasers without Inversion: Interference of Lifetime-Broadened Resonances," *Phys. Rev. Lett.* **62**, 1033 (1989).

12. S. E. Harris, J. E. Field, and A. Imamoglu, "Nonlinear Optical Processes Using Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **64**, 1107-1110 (March 1990).

13. K. H. Hahn, D. A. King, and E. E. Harris, "Nonlinear Generation of 104.8 nm Radiation Within an Absorption Window in Zinc," *Phys. Rev. Lett.* **65**, 2777-2779 (November 1990).

14. M. H. Sher, S. J. Benerofe, J. F. Young, and S. E. Harris, "2-Hz 109-nm Mirrorless Laser," *J. Opt. Soc. of Am.* **8**, 114-116 (January 1991).

15. J. D. Kmetec, J. J. Macklin, and J. F. Young, "0.5-TW, 125-fs Ti:Sapphire Laser," *Opt. Lett.* **16**, 1001-1003 (1991).

16. A. Imamoglu, J. E. Field, and S. E. Harris, "Lasers without Inversion: A Closed Lifetime Broadened System," *Phys. Rev. Lett.* **66**, 1154-1156 (March 1991).

17. K. -J. Boller, A. Imamoglu, and S. E. Harris, "Observation of Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **66**, 2593-2596 (May 1991).

18. S. J. Benerofe, Guang-Yu Yin, C. P. J. Barty, J. F. Young, and S. E. Harris, "116-nm H_2 Laser Pumped by a Traveling-Wave Photoionization Electron Source," *Phys. Rev. Lett.* **66**, 3136-3139 (June 1991).

19. A. Imamoglu, J. J. Macklin, and S. E. Harris, "Interference of Lifetime Broadened Resonances: Nonreciprocal Gain and Loss Profiles," in *New Frontiers in Quantum Electrodynamics and Quantum Optics*, A. O. Barut, ed. (Plenum Publishing Corp., New York, 1991), pp. 223-224.

20. C. P. J. Barty, G. Y. Yin, J. E. Field, S. J. Benerofe, J. F. Young, and S. E. Harris, "Photo-electron and Photoionization Pumping of XUV Lasers by Laser Produced Plasmas," in *X-ray Lasers*, G. J. Tallents, ed. (IOP Publishing Ltd., London, 1991), pp. 21-28.

21. J. E. Field, K. H. Hahn, and S. E. Harris, "Observation of Electromagnetically Induced Transparency in Collisionally Broadened Lead Vapor," *Phys Rev. Lett.* **67**, 3062-3065 (November 1991).

Section 6

Selected Abstracts

Laser-depletion spectroscopy of core-excited levels of neutral rubidium

J. K. Spong, A. Imamoḡlu, R. Buffa,* and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 6 May 1988)

This paper describes a new technique for obtaining level positions, linewidths, autoionizing times, and oscillator strengths of core-excited levels and transitions. A tunable laser is used to deplete the population of a radiating core-excited level, as other levels within the core-excited manifold are accessed. A saturation technique which allows the measurement of autoionizing times whose linewidth lies well beneath the combined Doppler-hyperfine profile is developed and demonstrated.

I. INTRODUCTION

Core-excited levels of atoms result from the excitation of an inner-shell electron to an outer, valence orbital. These configurations generally lie above the first continuum, and so are energetically allowed to autoionize. Knowledge of the locations and lifetimes of core-excited autoionizing levels is important to the understanding of many physical processes, including dielectronic recombination, multiphoton and multielectron ionization, and harmonic generation. Because these levels can be very energetic, they may also be applicable to the construction of extreme ultraviolet (xuv) lasers.[1] Traditionally, positions of core-excited levels have been measured by either ejected electron spectroscopy[2] or xuv photoabsorption spectroscopy[3] from the ground state. These techniques in general suffer from poor resolution and are therefore incapable of measuring autoionizing lifetimes which are longer than about 1 ps. More recently, Cooke *et al.*[4] and Bloomfield *et al.*[5] have used multistep and multiphoton excitation to prepare doubly excited column II atoms and to measure their linewidths and autoionizing times.[6]

In this article we describe a new technique called depletion spectroscopy,[7–9] which allows the measurement of levels positions, linewidths, autoionizing lifetimes, and transition oscillator strengths of core-excited levels to unprecedented accuracy. Autoionizing lifetimes can be measured in the range of 10^{-9}–10^{-14} sec, including lifetimes whose Lorentz widths are far narrower than the Doppler or hyperfine width of the transition. The technique is applied to the core-excited manifold of neutral Rb. Fifteen core-excited levels are located and identified, and oscillator strengths and autoionizing lifetimes are measured. Several of these levels are found to be surprisingly stable against autoionization, and radiate strongly in the xuv on previously unobserved transitions. The wavelengths and yields of the xuv radiation support the identifications and lifetime measurements reported here.

II. DEPLETION SPECTROSCOPY OF CORE-EXCITED LEVELS

To perform depletion spectroscopy, a radiating core-excited level is used as a reference level from which to access the autoionizing manifold, as illustrated in Fig. 1. The radiating level is impulsively excited, and a detector monitors the resulting fluorescence from this level. A tunable dye laser is passed through the excited vapor to transfer the fluorescing atoms to other core-excited levels nearby. As the laser is scanned in frequency, a level is encountered, and the excited level population is transferred to it, resulting in a depletion in the amount of fluorescence observed from the reference level. The location of the depletion signal, as a function of dye laser frequency, determines the energy of the accessed level, relative to the reference level. The shape of the depleted signal, as a function of laser intensity, can be analyzed to determine the oscillator strength and Lorentz width of the transition. Because the resolving instrument is the narrow bandwidth visible dye laser, the resolution of the technique is very high.

To apply depletion spectroscopy to the core-excited manifold of an element, a radiating, core-excited level must be available to serve as the reference level. Because core-excited levels tend to autoionize rapidly, states with acceptable fluorescent yields are extremely rare. Depletion spectroscopy is made possible by the recent identification of a subclass of core-excited levels, termed

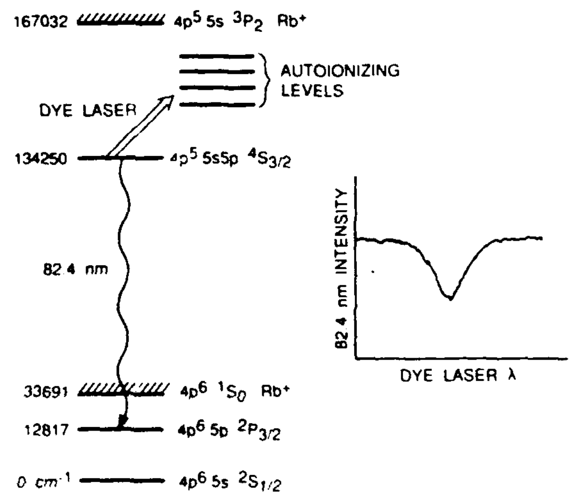

FIG. 1. Energy level diagram of Rb, showing the depletion spectroscopy technique.

Reprinted from Optics Letters, Vol. 14, page 168, February 1, 1989.

Extreme-ultraviolet fluorescence from core-excited levels of neutral rubidium

J. K. Spong, A. Imamoḡlu, J. D. Kmetec, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received July 5, 1988; accepted November 15, 1988

Strong extreme-ultraviolet fluorescence originating from core-excited autoionizing levels of neutral Rb is observed. Radiative branching ratios approaching unity are inferred from the radiative yields. Long autoionizing lifetimes and fast radiative rates make these levels promising candidates for extreme-ultraviolet laser systems.

In general, core-excited levels that lie above a continuum can decay by autoionization. Although there are exceptions in light elements for which a contradiction between orbital angular momentum and parity conservation may prohibit autoionization, in heavier elements the spin–orbit interaction leads to the breakdown of these selection rules. For these elements, excitation of electrons from the outermost closed shell results in levels that generally autoionize at least 100 times faster than they radiate. Because of their small radiative branching ratios, extreme-ultraviolet (XUV) fluorescence is seldom observed from such levels.

We recently reported the development of a tunable-laser-based technique that permits the measurement of long autoionizing times.[1,2] In the course of studying neutral Rb we measured lifetimes of 310 and >500 psec, respectively, for the $4p^5 5s6s\ ^2P_{1/2}$ and $^4P_{3/2}$ levels. Using the RCN/RCG atomic physics code,[3] we calculated radiative lifetimes of 8.3×10^{-10} and 2.9×10^{-9} sec for the XUV transitions at 75.7 and 76.2 nm ($4p^5 5s6s \rightarrow 4p^6 6s$), respectively, which originate from these levels. It was thus expected that we should be able to observe fluorescence on these transitions. The measurement of this fluorescence, together with an estimate of radiative branching ratios, is reported here.

The laser-induced fluorescence technique that we use is based on the properties of the Rb $4p^5 5s5p\ ^4S_{3/2}$ core-excited level. This level, termed quasi-metastable,[4,5] serves both as a reservoir of population and as an energy reference level.[6] Because of its 2.3% admixture with $4p^5 5s5p\ ^2P$, it radiates on the 82.4-nm $4p^5 5s5p\ ^4S_{3/2} \rightarrow 4p^6 5p\ ^2P_{3/2}$ transition and also permits laser coupling to odd-parity core-excited doublet and quartet levels.

The quasi-metastable level is excited by charge transfer from $Rb^+\ 4p^5 5s\ ^3P_1$ ions,[7] which are in turn produced by laser-generated x rays. As is shown in Fig. 1, a tunable dye laser is used to transfer quasi-metastable atoms to other, potentially radiating, levels in the core-excited manifold. As the laser is tuned through a transition, the quasi-metastable population is transferred to the radiating target level, resulting in a depletion of 82.4-nm fluorescence and the appearance of laser-induced fluorescence at a wavelength λ from the target level. The amount of laser-induced fluorescence is the intensity of the λ radiation with the laser tuned on line center, $I_\lambda^{(on)}$, minus the intensity with the laser tuned far off line center, $I_\lambda^{(off)}$. Similarly, the amount of laser-depleted fluorescence is the intensity at 82.4 nm with the laser tuned on line center, $I_{82.4}^{(on)}$, minus the intensity with the laser tuned off line center, $I_{82.4}^{(off)}$. The measured quantity in this experiment is the ratio of the laser-induced fluorescence to laser-depleted fluorescence:

$$R_i = \frac{I_\lambda^{(on)} - I_\lambda^{(off)}}{I_{82.4}^{(off)} - I_{82.4}^{(on)}}. \quad (1)$$

R_i is the relative fluorescent yield of the ith level and is used to infer the radiative branching ratio of the level, as described below. The uncertainty in the measurement of R_i is ±25%, and it is mainly due to the background noise of the signals.

The experimental apparatus is shown in Fig. 2. Soft x rays are produced by 150-mJ, 7-nsec pulses of 1.06-μm radiation focused through the Rb vapor onto

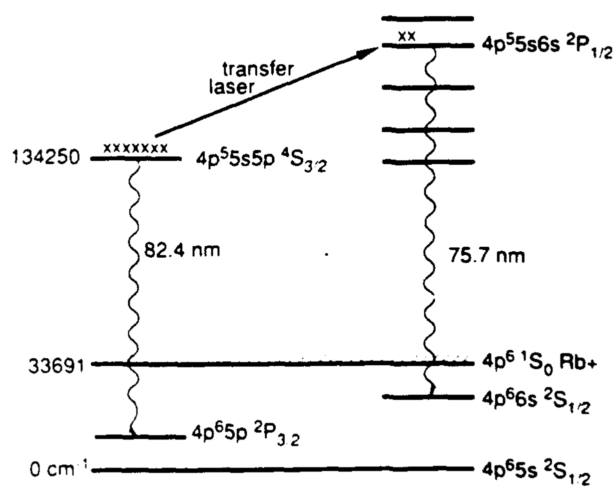

Fig. 1. Partial energy-level diagram of Rb showing the laser-induced fluorescence technique.

0146-9592/89/030168-03$2.00/0

12.8-eV Laser in Neutral Cesium

C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 20 July 1988)

We report the operation of a saturated 96.9-nm laser in Cs vapor that has an extrapolated small-signal gain of exp(83) in a total length of 17 cm. We believe that lasing occurs from a core-excited level embedded in the continuum of the valence electron. The laser is pumped by soft x rays from a synchronous, traveling-wave, laser-produced (2.5 J, 20 ps, 1064 nm) plasma.

PACS numbers: 42.55.Hq, 32.80.Dz, 42.55.Cj, 42.65.Re

In this Letter we report the operation of what we believe is the first laser with its upper level embedded in the continuum of the valence electron.[1] The laser employs a grating-assisted traveling-wave geometry[2,3] that creates a ~20-ps-long pulse of laser-produced soft x rays traveling synchronously with the generated 96.9-nm (12.8-eV) radiation. The gain coefficient is 4.9 cm^{-1} over a 17-cm length, which results in a total extrapolated small-signal gain of exp(83). After about 4 cm, the output energy grows linearly with length, indicating that the laser transition is fully saturated.

Core-excited levels that are embedded within a continuum usually autoionize rapidly, making the accumulation of population difficult. But this need not be the case; recent work by Spong *et al.*[4,5] has shown, for example, that there are many levels in neutral Rb that have autoionizing lifetimes exceeding 10 ps, and several that exceed 100 ps. Such long lifetimes can result either from angular momentum selection rules that to first order prohibit autoionization, or from fortuitous radial matrix element cancellations. The possibility of using such levels to make extreme ultraviolet and soft x-ray lasers has been noted by several workers.[6-8] The existence of an inversion from an upper level embedded within a continuum has been inferred from fluorescence intensity measurements by Silfvast and Wood.[9]

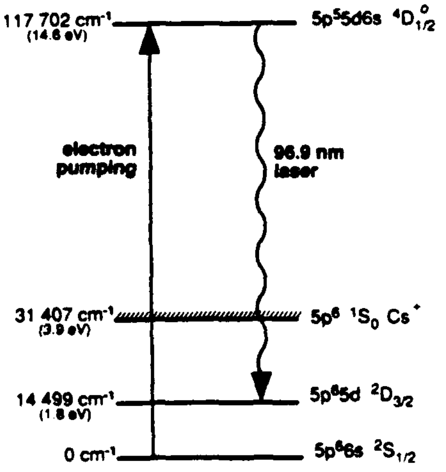

FIG. 1. Partial energy-level diagram for neutral Cs showing the laser transition.

Figure 1 is a partial energy-level diagram for the neutral Cs system showing the 96.9-nm laser transition. The 117702-cm^{-1} energy of the upper level has been measured by vacuum-ultraviolet absorption spectroscopy,[10,11] and the energy of the $5p^65d\,^2D_{3/2}$ lower level is well known.[11] The difference (96.897 nm) agrees with our measured emission wavelength of 96.86 ± 0.05 nm. Our identification of the upper level is based on a comparison of its characteristics as predicted by the multiconfigurational *RCN/RCG* atomic physics code of Cowan[12] with experimental measurements. Code-calculated oscillator strengths from the ground level are in good agreement with the absorption data of Connerade[10] and the ejected-electron data of Pejčev and Ross.[13] For simplicity, we have labeled the upper level $5p^55d6s\,^4D_{1/2}$ although it contains a large admixture of the $5d^2$ configuration. The code calculates a transition Einstein A rate of 2.3×10^7 s^{-1}, and an autoionizing rate of 1.6×10^{10} s^{-1}, yielding a radiative branching ratio of 0.0014. We calculate a Doppler-broadened stimulated emission cross section of 1.7×10^{-14} cm^2. Thus, it should be very difficult to observe spontaneous emission from this transition; significant outputs will occur only if the upper level is excited very rapidly and the stimulated emission rate exceeds the autoionizing rate. In our experiments rapid excitation is provided by the combination of a ~20-ps-long pumping pulse and a synchronous traveling-wave geometry.

The experimental geometry is a modification of that used by Sher *et al.*[2] for the Xe 108.9-nm Auger laser. As shown in Fig. 2, a 2.5-J, 15- to 20-ps-long 1064-nm pulse is incident upon a cylindrical lens at 65° from normal and is focused onto a target parallel to the lens. The width of the line focus is ~100 μm. The large angle of incidence expands the length of the line focus by $1/\cos65 = 2.4$, producing a 17-cm-long plasma. By itself, this geometry would produce a plasma sweeping along the target at a speed of $c/\sin65 = 1.1c$, resulting in a synchronism mismatch of 3.1 ps/cm of target length. In this experiment, however, the 20-ps-long pulse is formed

Lasers without Inversion: Interference of Lifetime-Broadened Resonances

S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 23 September 1988)

We show that if two upper levels of a four-level laser system are purely lifetime broadened, and decay to an identical continuum, then there will be an interference in the absorption profile of lower-level atoms, and that this interference is absent from the stimulated emission profile of the upper-level atoms. Laser amplification may then be obtained without inversion. Examples include interfering autoionizing levels, and tunneling systems.

PACS numbers: 42.55.Bi, 32.70.Jz, 32.80.Dz

While it is commonly believed that population inversion is a requirement for obtaining laser amplification, this is not so. In this Letter we show that if two upper levels of a four-level laser system are purely lifetime broadened, and decay to an identical continuum, then there will be an interference in the absorption profile of lower-level atoms, and this interference will be absent from the stimulated emission profile of the upper-level atoms. It is therefore possible to have a substantial gain cross section at frequencies where the absorption cross section is zero and to obtain laser amplification under conditions where the lower-level population greatly exceeds the upper-level population.

Figure 1 is an energy-level diagram for the analysis. Level $|1\rangle$ is the lower laser level. Levels $|2\rangle$ and $|3\rangle$ are upper levels which are assumed to be lifetime broadened and decay to the same continuum with decay rates Γ_2 and Γ_3. This decay may result, for example, from an Auger or autoionizing process. Level $|0\rangle$ is a reservoir from which the other levels may be excited by, for example, electron[1] or optical pumping. We consider two initial conditions: (a) atoms which at $t=0$ are in level $|1\rangle$, and (b) atoms which at $t=0$ are in level $|2\rangle$.

Those atoms which are initially in level $|1\rangle$ experience Fano-type interferences[2] in their absorption profile; i.e., there are several quantum-mechanical paths to the same final continuum level. Atoms which are initially in level $|2\rangle$ do not exhibit this type of destructive interference, but do display unusual properties. For example, even if the oscillator strength from level $|2\rangle$ to level $|1\rangle$ is zero, there will be gain on the $|2\rangle \rightarrow |1\rangle$ transition.

There is a precedent to this idea: Several years ago Arkhipkin and Heller[3] showed that a discrete level, which is embedded in a continuum and exhibits a Fano interference between photoionization to the continuum and (virtual) excitation of the discrete level, exhibits no such interference in emission. Here we show that the interference of two lifetime-broadened levels will create the same type of nonreciprocal interference. Though for generality we include a direct photoionization channel to the continuum, it is not an important or essential ingredient. The consideration of the interference of discrete lines allows much larger gain cross sections at the zero-loss point, and of more importance allows the application of these ideas to systems where the direct channel is absent or too small to be useful. Examples include systems where the lifetime broadening of the two upper levels occurs by autoionization, tunneling, or by radiative decay.[4]

The basis set for this work consists of three discrete levels and a density of continuum levels. We use a method described by Cohen-Tannoudji, Diu, and Laloe[5] and by Lambroupoulos and Zoller[6] to build the continuum into the coupled equations for the discrete levels. This is done by first integrating the continuum equations and then substituting the result back into the equations for the discrete levels. Thereafter, levels $|2\rangle$ and $|3\rangle$ are characterized by their decay rates to the continuum and their oscillator strengths, or equivalently by their Rabi frequencies to level $|1\rangle$. This method imposes a $t=0$, zero boundary condition on the continuum levels, and also requires that the matrix-element-weighted continuum be flat as compared to the spectral bandwidth of the

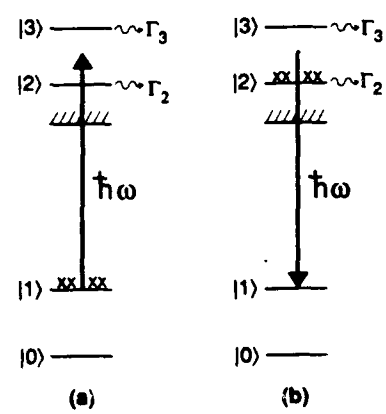

FIG. 1. Energy-level diagram for the analysis. (a) Atoms in level $|1\rangle$ at $t=0$ absorb probe radiation ($\hbar\omega$) and decay to an energy-conserving ion and electron. (b) Atoms in level $|2\rangle$ at $t=0$ both autoionize and are stimulated to level $|1\rangle$, thereby producing gain at the probe frequency. Levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ may be pumped by electrons or photons from level $|0\rangle$.

Non-Reciprocity of Autoionizing Interferences: Lasers Without Inversion

S. E. Harris

Edward L. Ginzton Laboratory, Stanford University
Stanford, California 94305

Abstract

Interferences of autoionizing lines may reduce or eliminate absorption of lower level atoms. Stimulated emission shows no such interferences, thereby allowing laser gain without population inversion.

Introduction

During the last several years we have made significant progress toward the realization of a class of extreme ultraviolet and soft x-ray lasers where the upper laser level is embedded in the continuum of the valence electron [1]. The advantage of this type of laser, as compared to more typical ionic lasers, is that the upper laser level occurs at much lower energy, and therefore may be pumped by cooler x-rays or electrons; this allows the operation of such lasers at pumping energies and powers which are much lower than would otherwise be possible. Highlights of recent work include: the demonstration that levels in the continuum may be pumped by electrons and retain their metastability under practical operating conditions [2]; the development of the technique of depletion spectroscopy for the measurement of autoionizing lifetimes [3]; and most recently the demonstration of an extraordinary 12.8 eV laser in neutral Cs [4]. This laser achieves an equivalent small signal gain of exp(83), at a pumping energy of 3 joules in 20 psec.

An energy level diagram for the Cs laser is shown in Fig. 1. Though we have a good understanding of the mechanism for pumping the upper level, we do not understand how inversion is obtained; that is, why do electrons not populate the lower laser level? Though earlier proposals [1] suggested the use of a ionizing laser to empty the lower level, no such laser was used in the Cs experiments.

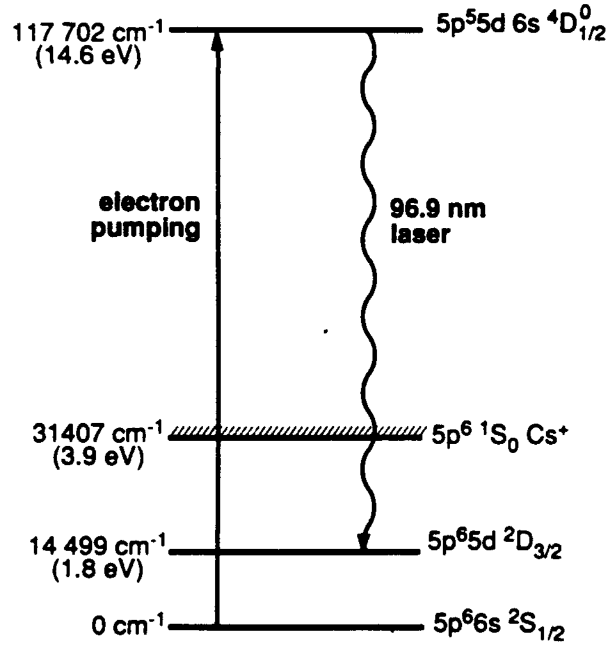

Figure 1—Partial energy level diagram for neutral Cs showing the laser transition.

In trying to understand this system the thought occured to me that perhaps inversion is not necessary. Perhaps, interference between two autoionizing levels could result in a cancellation of

12.8 eV Laser in Neutral Cesium

C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field,
J. F. Young, and S. E. Harris

Edward L. Ginzton Laboratory

Stanford University

Stanford, CA 94305

Abstract

We report the operation of a saturated 12.8 eV (96.9 nm) laser in Cs vapor that has an extrapolated small signal gain of exp(83) in a total length of 17 cm. We believe that lasing occurs from a core-excited level embedded in the continuum of the valence electron. The laser is pumped by soft x-rays from a synchronous, traveling-wave, laser-produced (2.5 J, 15 ps, 1064 nm) plasma.

Introduction

Recently, it has been the aim of several research efforts to achieve lasing action at wavelengths below 100 nm. In general, successful efforts such as the 20.6 nm Se laser[1] have involved transitions in highly ionized atoms created within a dense laser-produced plasma and have required kilojoule class laser systems as pumping sources. In this work, we report the first operation of a new class of short wavelength lasers in which the upper level of the lasing transition is embedded in the continuum of the valence electron[2]. Consequently, it requires less than a joule of pumping energy to saturate the lasing transition.

We believe that the upper level of the laser is a core-excited level in neutral Cs and is pumped by photoelectrons generated by soft x-rays emitted from a laser produced plasma. Core-excited levels that are embedded within a continuum usually autoionize on a picosecond timescale, making the accumulation of population difficult. But this need not be the case; recent work by Spong *et al.*[3,4] has shown, for example, that there are many levels in neutral Rb that have autoionization lifetimes exceeding 10 ps and several that exceed 100 ps. Such long lifetimes can result either from angular momentum and spin selection rules that to first order prohibit autoionization, or from fortuitous radial matrix element cancellations. The possibility of using such levels to make extreme ultraviolet and soft x-ray lasers has been noted by several workers[5,6,7]. The existence of an inversion from an upper level embedded within a continuum has been inferred from fluorescence intensity measurements by Silfvast et al.[8].

Spectroscopy

An energy level diagram of the Cs 96.9 nm laser system is shown in fig. 1. The 117,702 cm^{-1} energy of the upper level has been measured by vacuum ultraviolet absorption spectroscopy [9,10], and the energy of the $5p^6 5d$ $^2D_{3/2}$ lower level is well known[10]. The difference, 96.897 nm, agrees with our measured emission wavelength of 96.86 ± 0.05 nm. The upper level designation is

Targets for Efficient Femtosecond-Time-Scale X-Ray Generation

J.D. Kmetec and S.E. Harris

Edward L. Ginzton Laboratory, Stanford University,
Stanford, CA 94305

Abstract:

High power femtosecond lasers can be used to generate bright x-ray sources. The choice of element for the laser-produced-plasma target strongly affects the nature of the emitted x-rays. Combining low Z and high Z elements results in the brightest femtosecond x-ray radiators. Examples of lithium and tin combinations yielding 3.6 Å x-rays with up to several percent conversion efficiencies are given.

Introduction:

For many years, the output of pulsed laser systems have been focussed onto solid targets to generate plasmas. It is well known that a laser-produced- plasma can efficiently radiate in the XUV to x-ray regions of the spectrum. The recent development of very high peak power femtosecond lasers promises to yield even brighter plasmas emitting at even shorter wavelengths (1). We have studied the role of the laser target in a femtosecond-time-scale plasma and suggest several configurations which maximize the x-ray conversion efficiency while retaining the short pulse nature of the laser.

The dominant radiative processes in a laser-produced-plasma are line emission and two-body recombination. There have been experiments using picosecond and femtosecond lasers to drive x-ray-emitting plasmas (2-5), and also some measurements which show x-ray pulse lengthening well into the picosecond range (6). Both because of the relatively low power density on target, and because the targets were heavy metals, the electron temperatures were low, and the rise and fall times of the x-ray pulse were dominated by avalanche stripdown and two-body radiative recombination. Characteristic recombination times are slow compared to currently available femtosecond lasers. We propose conditions which maximize line radiation excited by hot electrons. The radiative rates and the heating and cooling times of the electrons can be much faster than recombination times.

The electrons are heated by the high power density of the laser, and cooled by inelastic ionizing collisions with the ions and by thermal diffusion. On a femtosecond time scale, ablation is negligible and thus very little cooling is achieved by plasma expansion. High Z elements have low thermal conductivity but very high inelastic cooling rates which prevent the laser from heating the electrons to high temperatures. Low Z elements quickly ionize completely and lose all inelastic

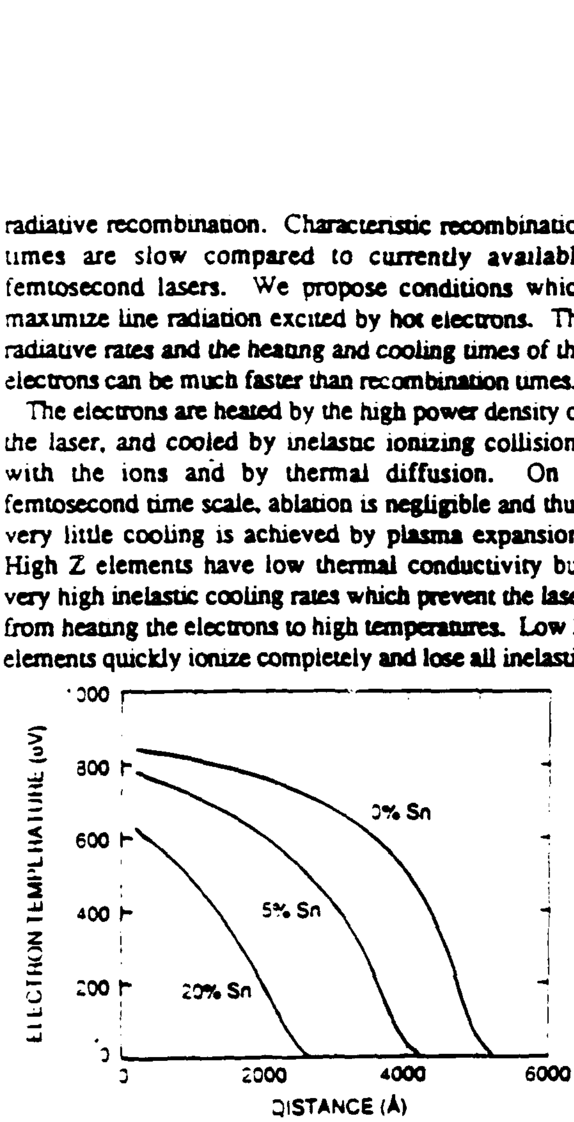

Figure 1: Electron temperature vs. distance for a solid Li-Sn mixed target at an absorbed energy of 1 kJ/cm^2, for different values of Sn density. The profiles are evaluated 50 fs after the peak of the laser pulse. Reproduced with permission from Ref. 7. Copyright 1988, American Physical Society.

Lasers without inversion: Single-atom transient response

S. E. Harris and J. J. Macklin
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 29 June 1989)

We discuss the effect of the transient response on the dynamics of lifetime broadened lasers that operate without the need for population inversion. A relationship between the steady-state absorptive transition probability rate and the transient gain and loss is given.

It has recently been shown that a population inversion is not a prerequisite for obtaining laser amplification and oscillation.[1-3] The essential idea is to utilize a system which causes a destructive interference in the absorption profile of lower-level atoms but not in the emission profile of upper-level atoms. An example of such a system is shown in the inset of Fig. 1(b). Level $|1\rangle$ is the lower laser level. Levels $|2\rangle$ and $|3\rangle$ are upper levels which are lifetime broadened with decay rates Γ_2 and Γ_3. This decay may result from autoionization, photoionization, tunneling, or spontaneous emission to a fourth level; but for the ideal case which we discuss here, the decay of both upper levels must occur to a common final continuum. (For example, for autoionization, this continuum is an ion and a free electron of prescribed angular momentum and arbitrary energy.)

In earlier work[1-3] the response of the lower-level atoms has been studied in the steady state, and the initial transient absorption caused by the excitation of the atoms at $t=0$ was ignored. Since this absorption effectively ends within several decay times of the upper levels, and since atoms may remain in level $|1\rangle$ indefinitely, the time-integrated contribution to the total absorption of this transient is vanishingly small, and is neglected when computing the steady-state (Fano-type) interference profiles of level $|1\rangle$ atoms. However, when operating at the zero of the interference profile, this transient absorption is the only absorption and should not be neglected. In fact, we show here, that this absorption determines what is, in effect, a threshold condition for lasers of this type.

When an atom is excited to level $|2\rangle$ or to level $|3\rangle$ (excitation occurs from some level or levels which are not shown), the stimulated response terminates within several decay times, and is, therefore, itself a transient response. For an ensemble of atoms, with rates into both upper and lower levels, the overall gain-loss balance is determined by the (single-atom) transient emission and the combined steady-state–transient absorption. It is often the case, for example in a cw discharge, that an ensemble of atoms is in steady state, while each individual atom has both a transient and a steady-state component. In this Rapid Communication we focus on the individual atom.

We use the equations of Ref. 1. These describe the threefold interference of level-$|1\rangle$ atoms to two upper levels and to the common continuum to which the upper levels decay. Results for a single level and a continuum, or two levels without a direct channel to the continuum, are obtained as special cases. The time-varying amplitudes of a lower level $|1\rangle$ and upper levels $|2\rangle$ and $|3\rangle$ are given by

$$\frac{\partial a_1}{\partial t}+j\Delta\tilde{\omega}_{11}a_1=\kappa_{12}a_2+\kappa_{13}a_3\,, \tag{1a}$$

$$\frac{\partial a_2}{\partial t}+j\Delta\tilde{\omega}_{21}a_2=\kappa_{12}a_1+\kappa_{23}a_3\,, \tag{1b}$$

$$\frac{\partial a_3}{\partial t}+j\Delta\tilde{\omega}_{31}a_3=\kappa_{13}a_1+\kappa_{23}a_2\,. \tag{1c}$$

The quantities in these equations are

$$\begin{aligned}
&\Delta\tilde{\omega}_{11}=-j\frac{W_C}{2}, \quad \kappa_{12}=\tfrac{1}{2}[j\Omega_{12}+(\Gamma_2W_c)^{1/2}],\\
&\Delta\tilde{\omega}_{21}=\Delta\omega_{21}-j\frac{\Gamma_2}{2}, \quad \kappa_{13}=\tfrac{1}{2}[j\Omega_{13}+(\Gamma_3W_c)^{1/2}]\,,\\
&\Delta\tilde{\omega}_{31}=\Delta\omega_{31}-j\frac{\Gamma_3}{2}, \quad \kappa_{23}=-\tfrac{1}{2}(\Gamma_2\Gamma_3)^{1/2},
\end{aligned} \tag{2}$$

where Γ_2 and Γ_3 are the decay rates of levels $|2\rangle$ and $|3\rangle$; $\Delta\omega_{21}=\omega_2-(\omega_1+\omega)$, $\Delta\omega_{31}=\omega_3-(\omega_1+\omega)$, and ω is the angular frequency of the electromagnetic field; Ω_{12} and Ω_{13} are the respective Rabi frequencies $(\mu E/\hbar)$, and W_c is the (direct channel) photoionization rate of level $|1\rangle$ to the continuum. We assume that the basis set has been prediagonalized[1] so that Γ_2, Γ_3, and W_c are real. We note the importance of the cross term κ_{23}, which represents the fact that as level $|2\rangle$ decays it drives level $|3\rangle$ and vice versa. This term arises since both levels couple to the same continuum level and therefore to each other.

We begin by examining computer solutions of these equations. The parameters for the computer runs (Fig. 1) are chosen so as to attain a zero in the steady-state absorption. The parameters for the three-level system are $\Omega_{12}=1/\sqrt{10}$, $\Omega_{13}=1$, $\Delta\omega_{21}=-5$, $\Delta\omega_{31}=50$, $\Gamma_2=1$, $\Gamma_3=10$, and $W_c=0$. For comparison, we also show a two-level system with the same parameters but without the interfering level $|3\rangle$.

Figure 1(a) shows the probability for level $|1\rangle$ occupancy, $|a_1(t)|^2$, versus time for the two-level system with the boundary condition $a_1(0)=1$, $a_2(0)=a_3(0)=0$. The absorption consists of the sum of a transient term and of a (golden-rule) steady-state term. Figure 1(b) shows this same quantity for the ideal three-level system. Here the steady-state term is zero (zero slope) and the absorption consists of only the transient term.

Figures 1(c) and 1(d) show the emission process. Here the boundary condition at $t=0$ is $a_2(0)=1$, $a_1(0)$

Nonreciprocal Emissive and Absorptive Processes

S. E. Harris, A. Imamoglu, and J. J. Macklin
Edward L. Ginzton Laboratory, Stanford University
Stanford, CA 94305, USA

1. Introduction

Though it has long been believed that an inversion is critical to obtaining laser amplification and oscillation, this is not the case. Recently, we have shown that if two levels decay to an identical continuum, that this decay couples these levels and results in nonreciprocal emissive and absorptive profiles (1,2). Though previous work (1-3) has emphasized systems where the decay results from autoionization or photoionization, these ideas also hold for purely radiative decay (4). Figure 1 shows such a system. Two upper states of the same angular momentum radiatively decay to the same final level and are coupled through this decay. In a sense, one may look at this coupling as an "internal radiative trapping." When a photon is spontaneously emitted by an upper state, it has a probability of being instantaneously reabsorbed by the other upper state of the same atom. It is in this sense that the upper states are coupled.

When one examines the absorption profile of atoms which are in state |1>, one finds interferences in the absorption profile - which in this case can be viewed as interferences in the anti-Stokes scattering profile. These interferences are not present in the emissive profile of atoms which at t=0 are in state |2> or |3>.

2. Non-Cancelable Channels

In previous work, we have studied the ideal case, where both upper levels decay to the same final state. There are always other non-cancelable channels which we have neglected. In particular there is always spontaneous emission on the laser channel itself. The assumption is therefore that the decay rate of the cancelable channel is much larger than the decay rate of the non-cancelable channels. For this condition, it may be shown that the ratio of the stimulated emission cross section to the absorption cross section is equal to the ratio of the cancelable and non-cancelable decay rates.

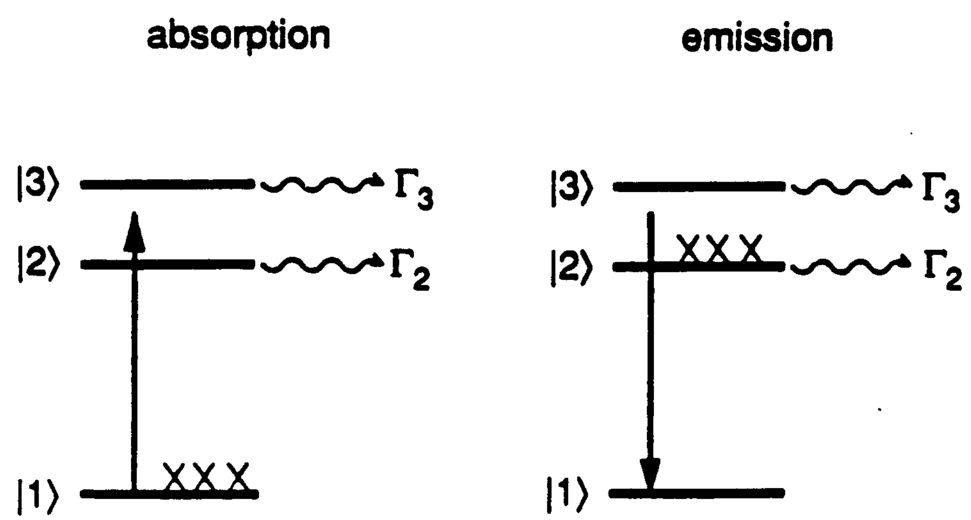

Fig. 1 Schematic of a system where the interference results from radiation broadening. The selection rule for this type of sytem is: states |2> and |3> must have the same parity, J, and m_J ; and must decay to a common final level (not shown in the figure).

Lasers without inversion: interference of dressed lifetime-broadened states

A. Imamoğlu and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received July 25, 1989; accepted October 4, 1989

We describe the use of a coupling electromagnetic field to provide a general method of producing inversion-free laser systems. The interference between dressed states produces a zero in absorption while allowing gains of the order of that of the uncoupled system.

It has recently been shown that if two upper states of a three-state laser system (Fig. 1) are purely lifetime broadened and decay to an identical continuum, this decay couples these states and results in nonreciprocal emission and absorption profiles.[1,2] One may obtain a zero in the absorption cross section and, for states that are spaced by several inverse lifetimes, obtain nearly the full gain cross section of a single transition.

The problem is that it is not easy to find nearby states that decay strictly to the same continuum. For states that decay by autoionization and Auger processes there are almost always several channels into which the state may decay. For states that decay by radiation, the decay rate is determined by interaction with the vacuum fluctuations, while their spacing is determined by electrostatic interactions; such states are therefore most often spaced by many inverse lifetimes.

In this Letter we show how to use an additional electromagnetic field to create a pair of interfering dressed states that *a priori* decay to the same continuum. Figure 2(a) shows the bare states and the electromagnetic field that is applied; Fig. 2(b) shows the equivalent dressed-state system. In the following we show that the effect of the interference of these dressed states is to create a zero in the absorption profile of state $|1\rangle$ atoms. In all cases this zero occurs at an energy that is the sum of the energies of (bare) state $|2\rangle$ and the coupling electromagnetic field. The emission profile of excited state $|2\rangle$ atoms does not exhibit this zero and, in fact, may have a gain cross section at the frequency of the zero that is of order of the gain cross section of the bare $|3\rangle$–$|1\rangle$ transition. This method thereby permits a general class of lifetime-broadened lasers that may operate without inversion.

In Fig. 2(a) we view the strength Ω_{23} and frequency ω_c of the coupling electromagnetic field as fixed and consider the gain and loss as a function of the probe frequency ω_p. We take the probe intensity Ω_{13} to be small as compared with Ω_{23} and Γ_3, where Γ_3 is the decay rate (to an arbitrary continuum) of state $|3\rangle$. The frequency detunings of the bare system are defined as $\Delta\omega_c = \omega_3 - \omega_2 - \omega_c$ and $\Delta\omega_p = \omega_3 - \omega_p - \omega_1$.

We transform to the equivalent dressed-state[3] system of Fig. 2(b). We assume that only a single pair of dressed states is in the vicinity of the probe frequency and that all other pairs may be neglected. The transformation from bare states $|2\rangle$ and $|3\rangle$ to dressed states $|2d\rangle$ and $|3d\rangle$ is

$$|2d\rangle = \cos\theta|2\rangle - \sin\theta|3\rangle,$$
$$|3d\rangle = \sin\theta|2\rangle + \cos\theta|3\rangle,$$
$$\tan 2\theta = \frac{-\Omega_{23}}{\Delta\omega_c}. \tag{1a}$$

The equivalent decay rates, Rabi frequencies, and detunings of the dressed system of Fig. 2(b) are then

$$\Gamma_{2d} = \Gamma_3 \sin^2\theta, \qquad \Gamma_{3d} = \Gamma_3 \cos^2\theta,$$
$$\Omega_{12d} = -\Omega_{13}\sin\theta, \qquad \Omega_{13d} = \Omega_{13}\cos\theta,$$
$$\Delta\omega_{2d} = \omega_2 - \frac{\delta}{2} + \omega_c - \omega_1 - \omega_p,$$
$$\Delta\omega_{3d} = \omega_3 + \frac{\delta}{2} - \omega_1 - \omega_p,$$
$$\delta = \frac{\Delta\omega_c(1 - \cos 2\theta)}{\cos 2\theta}. \tag{1b}$$

To obtain the absorption profile of the probe beam,

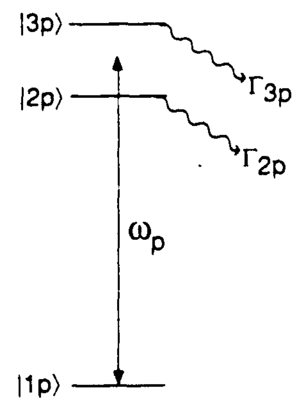

Fig. 1. Prototype system for inversion-free lasers. Γ_{2p} and Γ_{3p} are the decay rates, to the same continuum, of prototype states $|2p\rangle$ and $|3p\rangle$.

0146-9592/89/241344-03$2.00/0

2-Hz 109-nm mirrorless laser

M. H. Sher, S. J. Benerofe, J. F. Young, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received July 28, 1989; accepted June 8, 1990

We report 2-Hz operation of a single-pass 109-nm laser with a small-signal gain of exp 33 and a saturated output energy of 1 μJ. The laser is based on an oblique-incidence, laser-produced-plasma pumping geometry and requires only 500 mJ of 1064-nm energy in a 0.5-nsec pump pulse. We use the laser to produce a two-slit interference pattern and demonstrate a focusable intensity of greater than 10^9 W/cm^2.

We extended our oblique-incidence traveling-wave laser-produced-plasma pumping geometry[1] to 82.5° from normal (see Fig. 1). Using only 500 mJ of 1064-nm pump energy in a 0.5-nsec pulse, we observed a total small-signal gain of exp 33 and a saturated output energy of 1 μJ on the Xe III 109-nm Auger-laser transition.[2] Previous experiments on the Xe laser required significantly more pump energy to produce similar or smaller gains and were limited to extremely low repetition rates.[1–3] The present energy requirement makes it possible to run the 109-nm laser at the relatively high repetition rate of 2 Hz.

As was discussed in Ref. 1, the oblique-incidence grooved-target geometry has several advantages for efficient pumping of photoionization lasers. It produces a nearly synchronous, traveling-wave excitation source of a length that is determined by the angle of incidence. The grooved target brings the local angle of incidence of the beam-intercepting surfaces back to near normal, minimizing reflection and reducing the total length of the plasma. As a result, the pump-laser intensity on the target is independent of the angle of incidence, and the laser's efficiency of conversion to soft x rays is nominally independent of length.

The advantages of distributing the soft-x-ray source over an extended length can be understood qualitatively by using a simple model. The gain of the Xe laser, as well as of other photoionization lasers, is reduced by a high density of free electrons that are created in the pumping process. Thus, for a given Xe density and pump-pulse length, there is an optimum soft-x-ray flux that will result in a maximum gain coefficient. Because the soft-x-ray flux from a laser-produced plasma diverges, this gain coefficient will occur at some finite distance y above the surface of the target. If this distance is large compared with the groove spacing, the spatial variation of the flux from the series of small plasmas will be similar to that of a line source. As the angle of incidence is increased and the soft-x-ray energy is distributed over a longer length L, the maximum gain coefficient should remain constant as the region of optimum flux simply moves closer to the target as $1/L$. The total gain–length product will then be proportional to L. In addition, since both transverse dimensions of the maximum gain region will be proportional to y, and the laser output should be dominated by emission from this volume, a small increase in the length of the laser will produce a large improvement in the quality of the output beam. The laser-emission solid angle should scale as $1/L^4$, the Fresnel number as $1/L^3$, and the energy stored in the volume of maximum gain as $1/L$. One should therefore pump a photoionization laser at the maximum practical angle of incidence.

There are limits to this reasoning. The model breaks down if the pump energy per unit length is reduced to the point where y is of the order of the groove spacing or the transverse spot size. At this distance from the target the flux will no longer be constant in the longitudinal direction, and the line-source analogy will fail. Another restriction arises if the smallest dimension of the individual plasma spots, which is determined by both the angle of incidence and the groove spacing, becomes small enough to reduce the absorption of the pump laser and the soft-x-ray conversion efficiency.[4]

In order to test the predicted scaling of the gain–length product in Xe lasers, we compared the small-signal gain and large-signal behaviors that were produced at two different angles of incidence. A 2.1 cm × 1.0 cm elliptical 1064-nm laser beam (the smaller transverse dimension reduced the effect of aberrations in the tilted lenses and improved the focus) was used at $\theta = 67°$ and at $\theta = 82.5°$ to pump 5.3- and 16-cm-long lasers. The ratio of lengths and hence the predicted improvement in the gain–length product of the lasers was a factor of 3. The pump beam, which delivered 500 mJ of energy to the target in a 0.5-nsec pulse, was produced by a Nd:YAG laser system that was running at a repetition rate of 2 Hz. The cylindrical lenses for the 67° and 82.5° geometries had normal incidence focal lengths of 65 and 115 cm, respectively; the lens that was used at 82.5° had a specially developed antireflection coating[5] that transmitted 85% of the p-polarized light. The target was a stainless-steel tube, threaded at 31.5 grooves/cm, and the ambient Xe pressure was 10 Torr. We were able to hold several important pumping parameters constant by compensating for the slightly higher transmission of the lens used at 67° and by changing the groove angle of the target, α, from 45° to 60°. For both configurations the local angle of incidence $(\theta - \alpha)$ was 22°, the transverse spot size was 150 μm, and the intensity on target was 5×10^{10} W/cm^2. The longitudinal dimension of the individual plasmas, however, could not be held constant without changing the groove spacing.

0740-3224/91/010114-03$05.00

Nonlinear Optical Processes Using Electromagnetically Induced Transparency

S. E. Harris, J. E. Field, and A. Imamoğlu
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 27 December 1989)

We show that by applying a strong-coupling field between a metastable state and the upper state of an allowed transition to ground one may obtain a resonantly enhanced third-order susceptibility while at the same time inducing transparency of the media. An improvement in conversion efficiency and parametric gain, as compared to weak-coupling field behavior, of many orders of magnitude is predicted.

PACS numbers: 42.65.Ky, 42.50.Hz, 42.50.Qg

It is well known by those practicing the techniques of nonlinear optics that the power which may be generated in a frequency summing process, or the gain which may be obtained in a parametric process is determined by the interplay of the nonlinear and linear susceptibilities.[1,2] In general, as an atomic transition to the ground state is approached, the nonlinear susceptibility is resonantly enhanced, but at the same time the media exhibits a rapidly increasing refractive index and becomes opaque.

In this Letter we show how it is possible to create nonlinear media with resonantly enhanced nonlinear susceptibilities and at the same time induce transparency and a zero in the contribution of the resonance transition to the refractive index. An energy-level diagram for a prototype system is shown in Fig. 1. We apply a strong electromagnetic coupling field of frequency ω_c between a metastable state $|2\rangle$ and a lifetime-broadened state $|3\rangle$, and generate the sum frequency $\omega_d=\omega_a+\omega_b+\omega_c$. We assume that $|1\rangle$-$|3\rangle$ is a resonance transition and that in the absence of ω_c, radiation at ω_d is strongly absorbed.

When the Rabi frequency of the coupling field exceeds the Doppler width of the $|1\rangle$-$|3\rangle$ transition, the media becomes transparent on line center. This transparency occurs because of the destructive interference of the split (Autler-Townes) components of the $|1\rangle$-$|3\rangle$ transition. Though one might expect that this interference would also negate the nonlinearity that causes the generation of ω_d, this is not so; because of a sign change in the dressed eigenvectors, for generated frequencies lying between the Autler-Townes components, there is a constructive rather than a destructive interference in the nonlinear susceptibility.

Before proceeding we note earlier work: The use of electromagnetic fields to create transparency has been reviewed by Knight.[3] Armstrong and Wynne[4] observed that Fano-type interferences between photoionization and autoionization are not mirrored in $\chi^{(3)}$ profiles; Pavlov *et al.*[5] observed an enhancement of sum frequency generation by inducing a Fano-type state into the continuum. The work described here does not involve photoionization. State $|3\rangle$ may decay radiatively, or by autoionization, but if it decays by autoionization, then this work neglects the direct coupling of states $|1\rangle$ and $|2\rangle$ to the continuum.

In the following paragraphs we first consider the dressed susceptibilities of a single atom and thereafter include the effects of collisional and Doppler broadening.

We assume that an electromagnetic field with the frequencies ω_a, ω_b, ω_c, and ω_d is applied to the atom and calculate the total dipole moment at ω_d. This dipole moment may be expressed in terms of a linear and a third-order susceptibility. These susceptibilities depend on the magnitude of the coupling field ω_c and in this sense are dressed by the field. The pertinent quantities are defined as

$$E(t)=\mathrm{Re}\left\{\sum_{k=a}^{d}E(\omega_k)e^{j\omega_k t}\right\},\quad P(t)=\mathrm{Re}\left\{\sum_{k=a}^{d}P(\omega_k)e^{j\omega_k t}\right\},$$
$$P(\omega_d)=\epsilon_0\chi_D^{(1)}(-\omega_d,\omega_d)E(\omega_d)+\tfrac{1}{4}\epsilon_0\chi_D^{(3)}(-\omega_d,\omega_a,\omega_b,\omega_c)E(\omega_a)E(\omega_b)E(\omega_c). \tag{1}$$

The susceptibilities are calculated from the equations for the time-varying probability amplitudes of a single atom

$$\frac{db_1}{dt}=\frac{j\Omega_{12}}{2}b_2+\frac{j\Omega_{13}}{2}b_3,\quad \frac{db_2}{dt}+j\Delta\tilde\omega_{21}b_2=\frac{j\Omega_{12}^*}{2}b_1+\frac{j\Omega_{23}}{2}b_3,\quad \frac{db_3}{dt}+j\Delta\tilde\omega_{31}b_3=\frac{j\Omega_{13}^*}{2}b_1+\frac{j\Omega_{23}^*}{2}b_2, \tag{2a}$$

$$\Delta\tilde\omega_{21}=\Delta\omega_{21}-\frac{j\Gamma_2}{2},\quad \Delta\tilde\omega_{31}=\Delta\omega_{31}-\frac{j\Gamma_3}{2},\quad \Omega_{12}=\sum_i\frac{\Omega_{1i}\Omega_{i2}}{2}\left[\frac{1}{\omega_i-\omega_a}+\frac{1}{\omega_i-\omega_b}\right]. \tag{2b}$$

The Ω_{ij} are the respective Rabi frequencies; Ω_{12} is an effective Rabi frequency which is obtained by summing over in-

Lasers without Inversion: A Closed Lifetime Broadened System

A. Imamoḡlu, J. E. Field, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 21 September 1990)

We show a model laser system which operates by an electromagnetically induced interference. Provided that an inversion condition for the thermal radiation field is satisfied, the system lases without atomic population inversion in steady state. The system is pumped by incoherent radiation on the transition on which lasing occurs.

PACS numbers: 42.50.Hz, 32.80.Bx, 42.55.Bi

Recently, there has been considerable interest in lasers that operate without the need for a population inversion. These systems are based on a quantum interference effect which in atomic systems that are dominantly lifetime broadened creates different emissive and absorptive profiles.[1-11] Two related questions[8,9] have drawn attention: (1) Is there a hidden atomic basis set in which the system is inverted? (2) Must such systems be pumped from an external state which, like Raman lasers, is inverted with regard to the lower laser state?

In this Letter, we address these questions by proposing a model system (Fig. 1), which is closed with regard to the atomic basis set: That is, the lifetime broadening occurs by radiative decay to other states within the same system, and the laser is pumped by incoherent photons from its lower state. This closed system lases without atomic population inversion in steady state, provided that the occupancy of the radiation field modes satisfies an "inversion" condition.

Before proceeding further we note that Kocharovskaya and co-workers[12] and Scully and co-workers[13] have proposed a different class of laser systems which are based on a phase-dependent quantum interference. Such systems do not depend on lifetime broadening and the considerations of this work may not apply.

In the system of Fig. 1, following Imamoḡlu and Harris,[7] a strong electromagnetic field of frequency ω_c couples a metastable state $|2\rangle$ to a state $|3\rangle$ which decays radiatively to states $|2\rangle$ and $|1\rangle$. This coupling field creates a pair of dressed states which *a priori* decay to the same final states. The absorption of state $|1\rangle$ atoms as measured by a probe of frequency ω_p will exhibit a destructive interference with a minimum absorption at the frequency $\omega_2+\omega_c-\omega_1$. There are two essential requirements for the system: (i) The spontaneous decay rate from state $|3\rangle$ to state $|2\rangle$ exceeds that from state $|3\rangle$ to state $|1\rangle$, (ii) the average number of thermal photons per mode in the $|1\rangle$-$|3\rangle$ channel exceeds that in the $|2\rangle$-$|3\rangle$ channel.

We have, in part, chosen the system of Fig. 1 since all pumping and decay processes are radiative and therefore the density matrix for the system may be derived without the addition of phenomenological terms. We take the system to be pumped by incoherent radiation which has a different intensity (temperature) on the $|1\rangle$-$|3\rangle$ and on the $|2\rangle$-$|3\rangle$ transitions and which at each transition is spectrally broad as compared to the larger of the decay rate Γ_{32}, detuning $|\omega_2+\omega_c-\omega_3|$, or the Rabi frequency $|\Omega_{23}|$. We assume that the effect of the atoms on the pumping radiation is negligible and that each laser field is represented by a single coherent state. The derivation also assumes the Rabi frequencies to be much smaller than the corresponding laser frequencies. By tracing the global density matrix over the radiation field variables we obtain the equation of motion for the atomic density matrix elements in a frame rotating at the probe frequency ω_p.[14] These equations are the following:

$$\frac{d\rho_{11}}{dt}=\Gamma_{31}\rho_{33}+\tfrac{1}{2}i\Omega_{13}(\rho_{31}-\rho_{13})+R_{13}(\rho_{33}-\rho_{11}), \tag{1a}$$

$$\frac{d\rho_{22}}{dt}=\Gamma_{32}\rho_{33}+\tfrac{1}{2}i\Omega_{23}(\rho_{32}-\rho_{23})+R_{23}(\rho_{33}-\rho_{22}), \tag{1b}$$

$$\frac{d\rho_{33}}{dt}=-(\Gamma_{32}+\Gamma_{31})\rho_{33}+\tfrac{1}{2}i\Omega_{13}(\rho_{13}-\rho_{31})+\tfrac{1}{2}i\Omega_{23}(\rho_{23}-\rho_{32})-R_{23}(\rho_{33}-\rho_{22})-R_{13}(\rho_{33}-\rho_{11}), \tag{1c}$$

$$\frac{d\rho_{12}}{dt}=-(\tfrac{1}{2}\gamma_{21}-i\Delta\omega_{21})\rho_{12}-\tfrac{1}{2}i\Omega_{23}\rho_{13}+\tfrac{1}{2}i\Omega_{13}\rho_{32}, \tag{1d}$$

$$\frac{d\rho_{13}}{dt}=-(\tfrac{1}{2}\gamma_{31}-i\Delta\omega_{31})\rho_{13}+\tfrac{1}{2}i\Omega_{13}(\rho_{33}-\rho_{11})-\tfrac{1}{2}i\Omega_{23}\rho_{12}, \tag{1e}$$

$$\frac{d\rho_{23}}{dt}=-(\tfrac{1}{2}\gamma_{32}-i[\Delta\omega_{31}-\Delta\omega_{21}])\rho_{23}+\tfrac{1}{2}i\Omega_{23}(\rho_{33}-\rho_{22})-\tfrac{1}{2}i\Omega_{13}\rho_{21}, \tag{1f}$$

116-nm H_2 Laser Pumped by a Traveling-Wave Photoionization Electron Source

S. J. Benerofe, Guang-Yu Yin, C. P. J. Barty, J. F. Young,[a] and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 4 October 1990)

We report the use of a photoionization electron source to pump a 116-nm laser in the Werner band ($C\,^1\Pi_u \rightarrow X\,^1\Sigma_g^+$) of molecular hydrogen. The laser is pumped by free electrons which are created by photoionizing molecular hydrogen with soft x rays from a traveling-wave laser plasma. We show that even though the free electrons have an average temperature of ~10 eV, the lasing hydrogen molecules retain an ambient temperature of ~0.01 eV. This allows an extrapolated small-signal gain of exp(43), with a 1064-nm pumping energy of 580 mJ in 200 psec.

PACS numbers: 52.50.Jm, 34.50.Gb, 42.55.Em

We describe the use of a traveling-wave photoionization electron source[1] (PES) to pump a 116-nm laser in the Werner band of H_2. The PES is constructed by using a grazing-incidence, traveling-wave laser plasma[2] to make soft x rays which in turn photoionize ambient hydrogen molecules (Fig. 1). The electrons have an average energy which corresponds to the difference in energy of the pumping x rays and the ionization potential of H_2, and at sufficient pumping intensity may have a density which corresponds to a discharge current in excess of MA/cm^2. The rise time of the electron density is equal to that of the x-ray source, and may be picoseconds or shorter in duration, making PES an ideal source for pumping short-wavelength lasers.[3]

In this Letter we quantitatively demonstrate the advantages of PES over conventional electron pumping sources by generating saturated laser emission at 116 nm in the Werner band of H_2. We emphasize a special feature of this type of excitation, which is its ability to produce hot electrons while at the same time retaining an ambient (lasing medium) temperature which is comparatively cold. This is confirmed by measurements of the 116-nm gain as a function of the ambient H_2 temperature. In this work the free electrons have an average temperature of 10 eV while the lasing hydrogen molecules retain an ambient temperature of ~0.01 eV. This allows very high gain at modest pumping energy; here we obtain an extrapolated small-signal gain of exp(43), with a 1064-nm pumping energy of 580 mJ in a pulse width of 200 psec.

Before proceeding we note that previous short-wavelength lasers have operated in the focus of the incident laser and under conditions where the exciting electrons and target ions are relatively thermalized.[4] H_2 lasers have been constructed by using a Blumlein discharge (Waynant)[5] and by using a field-emission diode (Hodgson and Dreyfus).[6] In this work we obtain a gain coefficient which is over an order of magnitude larger than that previously obtained, demonstrating quantitatively one of the advantages of PES over conventional electron pumping sources.

Using a simple model for the PES excitation mechanism we will verify that in the present experiment the H_2 laser is pumped by electrons and is not directly photopumped. We will then proceed to describe the experimental setup and results. The calculation of the gain for the 116-nm laser pumped by a PES proceeds as follows: The 580-mJ, 200-psec, 1064-nm pump laser pulse creates a plasma on the target surface. Following Ref. 1 the spectral distribution of the soft x rays from the laser-produced plasma is modeled as a blackbody [see Fig. 1(a)] which has a characteristic temperature determined by the conversion efficiency of the pump laser to soft x rays and a pulse width comparable to that of the pump laser. From previous measurements of conversion efficiency done under similar experimental conditions,[7] we estimate the conversion efficiency of our laser to be approximately 2% into the energy range of interest. This implies a blackbody temperature of 12 eV. The soft x rays photoionize some of the hydrogen molecules surrounding the target, creating free electrons. In Fig. 1(b) we show the photoionization cross section for H_2.[8] Combining the blackbody-flux spectral distribution and the photoionization cross section, we calculate an electron density of 3×10^{15} cm^{-3} in the lasing region, and an average electron energy of 10 eV [Fig. 1(c)]. This is equivalent to a discharge current of about 9×10^4 A/cm^2. The cross section for electron excitation of the Werner band has been calculated by Gerhart[9] and is shown in Fig. 1(d). The cross section for pumping the 116-nm line is then calculated using Refs. 10 and 11. By multiplying the electron distribution by the electron pumping cross section for the upper laser level, and integrating, we estimate an upper-state ($C\,^1\Pi_u$, $v'=1$, $J'=1$) density of approximately 6×10^{12} cm^{-3} at room temperature [note that the upper laser level has a lifetime of approximately 600 psec (Ref. 12)]. This yields a calculated gain on the 116-nm transition of 0.35 cm^{-1} at room temperature (293 K).

This PES gain calculation is to be compared to the gain calculated for direct photopumping of the upper laser level. For this calculation we use the 12-eV black-

Nonlinear Generation of 104.8-nm Radiation within an Absorption Window in Zinc

K. H. Hahn, D. A. King, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 3 August 1990)

Two autoionizing levels which are separated by a few decay widths may exhibit a sharp interference or window in their absorption profile and also make canceling contributions to the refractive index at the absorption minimum. A correct choice of intermediate mixing levels prevents a similar cancellation in the nonlinear susceptibility. Using UV lasers with energies of about mJ and pulse lengths of 5 ns, we generate 0.23 μJ per pulse at 104.8 nm.

PACS numbers: 42.65.Ma, 32.80.Dz, 42.65.Ky

On occasion two atomic energy levels which lie above an ionization potential and decay by autoionization to the same continuum may exhibit a strong destructive interference in their absorption profile. In this Letter we show how, by the correct choice of intermediate mixing levels, one may use such a window to generate sum-frequency radiation.

Before proceeding we note that Armstrong and Wynne[1] have shown that the profiles for absorption and for the nonlinear susceptibility $\chi^{(3)}$ in the vicinity of a single autoionizing resonance need not be the same. By utilizing the interference of two lines, rather than that of a single line and a continuum, this work extends theirs in two ways: First, the interference of two lines results in a near zero in the contribution of the lines to the refractive index at the generated frequency, as well as to the absorption. This is not an accident and will occur whenever autoionization is determined by the same dipole matrix elements that determine the absorption. Second, because the interference between two lines is much steeper than that of a single line and a continuum, it occurs closer to both lines and results in a much greater increase in the nonlinearity length product.

The connection to the suggestion of Harris, Field, and Imamoğlu[2] and to the recent experiments of Hakuta, Marmet, and Stoicheff[3] is also to be noted: By applying an electromagnetic field to mix a (perfectly) metastable level with a lifetime-broadened level, one may create (in the absence of Doppler and collision broadening) the ideal situation of zero loss, zero contribution to the refractive index, and constructive interference in the nonlinearity. This work does not quite accomplish these goals: Here, the nonlinear susceptibility and the small but nonzero linear susceptibility are the result of the departure from L-S coupling.

Figure 1 shows a partial energy-level diagram of neutral Zn. The interfering levels $3d^9 4s^2 4p\,^1P_1^\circ$ and $3d^9 4s^2 4p\,^3D_1^\circ$ decay primarily by autoionization to a common $\mathrm{Zn}(3d^{10}4s\,^2S_{1/2})$ ion and a $^1P_1$ electron. Figure 2 shows the absorption cross section as a function of photon frequency, as obtained from the data of Marr and Austin.[4] The absorption window near 95 370 cm^{-1}

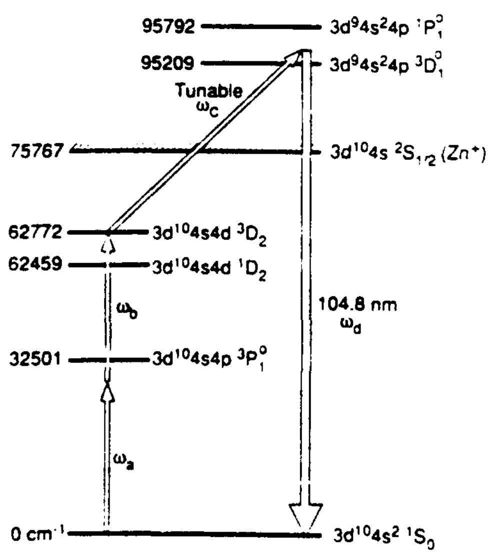

FIG. 1. Partial energy-level diagram of neutral zinc.

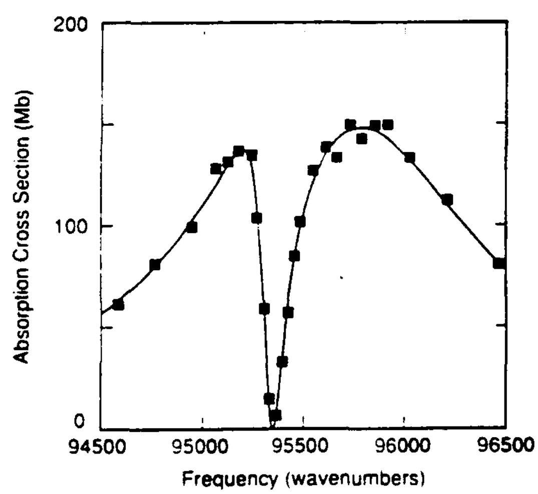

FIG. 2. Absorption cross section as a function of frequency. Data points are from Marr and Austin (Ref. 4). The solid curve is a fit using Eq. (6) from Harris (Ref. 5).

INTERFERENCE OF LIFETIME BROADENED RESONANCES: NONRECIPROCAL GAIN AND LOSS PROFILES

A. Imamoğlu, J. J. Macklin, and S. E. Harris

Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305

INTRODUCTION

It is commonly believed that laser systems have reciprocal absorption and emission profiles, and that population inversion is a necessary condition for obtaining laser amplification. Recently, it has been shown that interferences in lifetime broadened systems can result in nonreciprocal absorption and emission profiles.[1,2] At frequencies where the absorption rate goes to zero, light amplification is possible even though the lower level population greatly exceeds the upper level population. Figure 1 shows a prototype system: The two upper states $|2\rangle$ and $|3\rangle$ must have the same parity, total angular momentum J, and m_j quantum numbers;[3] and must decay to an identical continuum. The decay could be due to spontaneous emission to the states of a final level $|f\rangle$ (not shown in figure) or due to autoionization. The steady state loss mechanism for atoms initially in state $|1\rangle$ is Raman scattering (or virtual autoionization) into a final continuum. This process takes place through two intermediate states, $|2\rangle$ and $|3\rangle$, and is subject to quantum interference effects. The emission profile of an atom initially excited into state $|2\rangle$ or $|3\rangle$ from a reservoir does not show such interference effects. We have shown that these results also hold for two dressed states produced by using an additional electromagnetic field.[4] Selection rules simplify for such a system, making it possible to make lasers that operate without inversion.

ANALYSIS OF A RADIATIVELY-BROADENED SYSTEM

The basis set that we use in the analysis consists of the eigenstates of the non-interacting atom plus radiation field Hamiltonian. Assuming no photons in any but the laser mode of the radiation field initially, we can write the state vector of the total system in the interaction representation as[3,5]

Observation of Electromagnetically Induced Transparency

K.-J. Boller, A. Imamoğlu, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 12 December 1990)

We report the first demonstration of a technique by which an optically thick medium may be rendered transparent. The transparency results from a destructive interference of two dressed states which are created by applying a temporally smooth coupling laser between a bound state of an atom and the upper state of the transition which is to be made transparent. The transmittance of an autoionizing (ultraviolet) transition in Sr is changed from exp(−20) without a coupling laser present to exp(−1) in the presence of a coupling laser.

PACS numbers: 42.50.Qg, 32.80.Dz, 42.50.Hz, 42.65.Ky

This Letter reports the results of an experiment showing how an opaque atomic transition may be rendered transparent to radiation at its resonance frequency. This is accomplished by applying an electromagnetic coupling field between the upper state $|3\rangle$ of the transition and another state $|2\rangle$ of the atom (Fig. 1). When the Rabi frequency of the coupling field exceeds the inhomogeneous width of the $|1\rangle$-$|2\rangle$ transition, the medium becomes transparent on line center. In this work, the transmittance of an ultraviolet transition to an autoionizing state of neutral Sr is changed from exp(−20) without a coupling laser present to exp(−1) in the presence of a coupling laser.

The transparency described here is not a saturation or hole-burning phenomena. As the probe laser is turned on in the presence of the coupling laser, ground-state atoms evolve to a steady state where a fraction (in these experiments, about one part in a thousand) of their population is in state $|2\rangle$. This coherently phased population produces a dipole moment of equal magnitude and opposite sign to the primary dipole moment at the probe frequency. Equivalently, the transparency may be viewed as resulting from a combination of the ac-Stark splitting and *the interference between* the two dressed states which are created by the coupling laser [Fig. 1 (inset)]. It is the observation of the large change in opacity which results from this interference that is the special feature of this work.

There has been substantial theoretical work on laser-induced and laser-inhibited autoionization and the transparency observed here is a special case of the formalism and examples given by Lambropoulos,[1] Eberly,[2] Agarwal,[3] and their co-workers. The related subject of laser-induced continuum structure has been reviewed by Knight, Lauder, and Dalton.[4] Experimentally,[5] resonances induced into a continuum by a laser have been shown to cause a polarization rotation, to increase the efficiency of third-harmonic generation, and to modify three-photon ionization. An interference in autoionization as caused by a dc electric field has been described. This interference is also of the same nature as that observed by Whitley and Stroud[6] and Alzetta *et al.*[6] in the Rayleigh scattering of radiatively broadened systems.

Figure 1 shows a partial energy-level diagram of neutral strontium.[7] The probe transition which is to be rendered transparent is $5s5p\,^1P_1$-$4d5d\,^1D_2$ with a transition wavelength of 337.1 nm. The absorption oscillator strength of this transition, as calculated from the *RCN-RCG* atomic physics code,[8] is $gf = 0.15$. The upper level of this transition decays by autoionization and has a measured width of 1.2 cm^{-1}, corresponding to an autoionizing time of 4.4 psec. Its lower level is populated from ground by using a resonant 460.7-nm pulsed dye

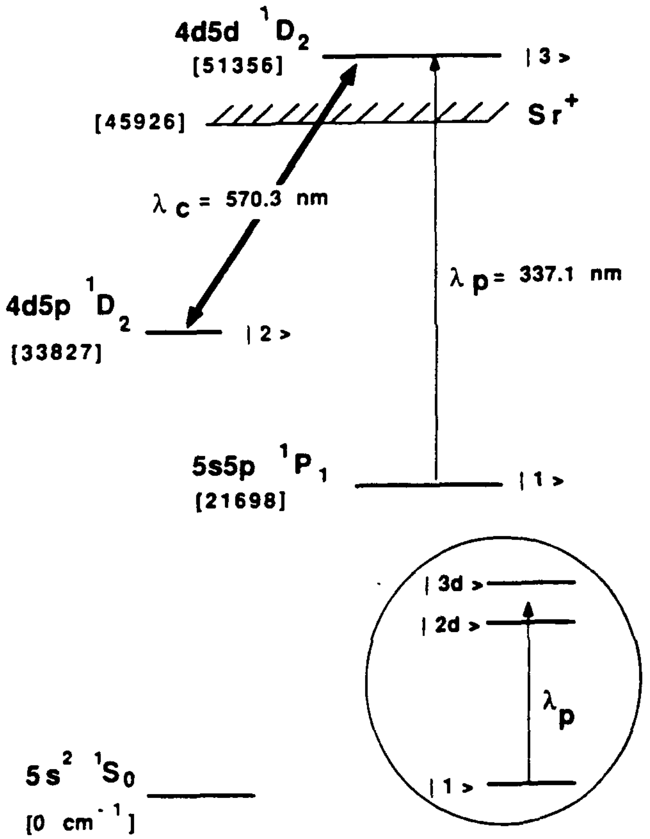

FIG. 1. Energy-level diagram of neutral Sr. Inset: Dressed-state picture.

Inst. Phys. Conf. Ser. No 116
Paper presented at Int. Colloquium on X-ray Lasers, York

Photo-electron and photoionization pumping of XUV lasers by laser produced plasmas

C P J Barty, G Y Yin, J E Field, S J Benerofe, J F Young and S E Harris

Edward L Ginzton Laboratory, Stanford University, Stanford CA 94305

ABSTRACT: Experiments involving two different classes of laser produced plasma pumped XUV lasers are presented. In photoionization pumped systems, preferentially ionization of inner shell electrons leads directly to inversion. In photo-electron pumped systems, electrons created by photoionization collisionally excite a neutral species. Photoionization pumped lasers in Xe, Kr, Zn, Cs, and K and photo-electron pumped lasers in Cs and H_2 are reviewed.

1. PHOTOIONIZATION PUMPED LASERS

The use of photoionization to create an inversion in the XUV was first suggested by Duguay and Rentzepis (1967). Their proposal was based on the photoionization of inner shell electrons in atomic Na. For photon energies greater than 36 eV, the photoionization cross section of a 2p electron in Na is roughly 100 times that of an outer 3s electron. Thus ionization by high energy photons will create an inversion on the 37.2 nm resonance transition of the first ion. However, the 3s electron of Na has a much larger electron ionization cross section and therefore the photo-electron created by the pumping process may produce ground state ions and thus destroy the inversion. This laser has never been successfully demonstrated. Since 1967, there have been numerous photoionization-pumped laser suggestions (Mani et al 1976, McGuire 1975, Axelrod 1976, Mendelsohn and Harris 1985, Walker et al 1986, Kapteyn et al 1986, Caro et al 1986), which have either directly or indirectly addressed problems present in the 37.2 nm Na II system.

One method of circumventing electron quenching problems is to utilize systems in which the lower level of the laser transition can not be directly accessed by electron collisions. The 108.9 nm Xe III Auger laser has become the most successful example of such a scheme. The Xe III system was first proposed and demonstrated by Kapteyn et al (1986). For photon energies greater than 67 eV, the largest photoionization cross section in Xe is for the removal of a 4d electron. Photoionization by soft x-rays from a high temperature laser-produced plasma may therefore result in a large density of Xe II, $4d^9 5s^2 5p^6\ ^2D_{3/2,5/2}$, ions. These ions lie 34 eV above the first continuum of the $4d^{10}5s^2 5p^4$ configuration of Xe II and are thus energetically unstable. As a result, they rapidly undergo Auger decay into various configurations of Xe III. Inversion occurs between the $4d^{10}5s^0 5p^6\ ^1S_0$ level of Xe III and the $4d^{10}5s^1 5p^5\ ^1P_1$ level 10 eV below. The transition has a Doppler-broadened gain cross section of 3×10^{-13} cm^2. Note that unlike the

0.5-TW, 125-fs Ti:sapphire laser

J. D. Kmetec, J. J. Macklin, and J. F. Young*

Edwa.d L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received February 6, 1991

We describe a terawatt, femtosecond laser system based on Ti:sapphire amplifiers and the technique of chirped-pulse amplification. The laser output at 807 nm contains 60 mJ of energy in a 125-fs pulse duration. The laser beam is nearly diffraction limited and may be focused to an intensity of 10^{18} W/cm^2 with an $f/6$ optic.

We describe a terawatt, femtosecond laser system based on titanium-doped-sapphire amplifiers and the technique of chirped-pulse amplification.[1-3] This technique uses group-velocity dispersion to expand and recompress the laser pulse width and permits a significant reduction in laser power in the amplifier. This process reduces the intensity-dependent pulse distortion that would otherwise limit the amplifier output energy. Previous research employing large-bandwidth amplifiers such as Ti:sapphire[4,5] and alexandrite[6] have obtained femtosecond pulses at the millijoule level. We report the generation of femtosecond pulses with 60 mJ of energy, while maintaining low distortion of the spatial and temporal properties of the output beam. In our system a positively dispersive grating pair stretches the 85-fs pulses from a mode-locked dye laser to 180 ps. These pulses seed a Ti:sapphire regenerative amplifier, which produces 8-mJ, 180-ps pulses at ~6 Hz. Further amplification in a triple-passed Ti:sapphire amplifier increases the pulse energy to 110 mJ. A negatively dispersive parallel grating pair then removes the chirp and thus recompresses the pulse width to 125 fs. The output contains 60 mJ of energy and a power of 0.5 TW. The spatial beam profile is Gaussian and measured to be 1.2× the diffraction limit. The low distortion of the pulse in space and time may enable one to obtain a focused intensity of 10^{18} W/cm^2 by using an $f/6$ optic. Prepulse power due to amplified spontaneous emission (ASE) is reduced to 10^{-9} of the peak power with the use of a saturable absorber, with a small decrease in beam quality. Only a single, commercially available, pump laser is required for all amplification.

The laser system is shown in Fig. 1. The femtosecond oscillator for the system is a hybridly mode-locked dye laser[7] operated at 807 nm. The gain and saturable absorber dyes are LDS-751 and HITC-I. The gain dye is pumped with 70-ps pulses of frequency-doubled light from a cw mode-locked Nd:YAG laser. The average output power of the dye laser is 5 mW, with a pulse width of 85 fs and a bandwidth of 8 nm. It is important to ensure that these seed pulses have sufficient bandwidth and contain no satellite pulses when one uses chirped-pulse amplification; otherwise damage may occur in the amplifier components. To provide reproducibly short, satellite-free pulses, we employ stabilization of the pump laser amplitude, use a birefringent filter to restrict wavelength and control bandwidth, and actively stabilize the dye-laser cavity length. In addition, the dye-laser cavity length is detuned by 1 μm from the pump frequency, a process that produces a periodic modulation of the output in time, whereas, for a large fraction of the period, the dye laser cannot generate satellite pulses. Since the occurrence of the satellite pulses is periodic and therefore predictable, we can reliably gate the amplifiers off during these times and so protect the amplifier components from damage.

The dye-laser output is collimated and double passed through an antiparallel grating pair[2,8] containing a 1:1 telescope that is necessary to produce positive group-velocity dispersion. This arrangement introduces a large, positive chirp to the pulse and stretches the pulse width by ~2000 times. The expander gratings are gold-coated, 1800-lines/mm holographic diffraction gratings, separated by 128 cm. The incident and diffracted angles for the first grating are 54.9° and 38.9°. Two 50-cm focal-length lenses, placed a distance $2f$ apart, form the telescope between the gratings. The lenses are cemented doublets and exhibit a $df/d\lambda = 0.01$ mm/nm at 800 nm. The effective length of the expander is 68 cm. The stretched pulse width, measured with a single-shot streak camera, is typically 180 ps, and the pulse energy output is 5 pJ.

To obtain a gain of 10^{10}, we use a regenerative amplifier and a triple-passed power amplifier employing Ti:sapphire, both antireflection coated with a single layer of MgF_2. The frequency-doubled output from a Q-switched, 10-Hz Nd:YAG laser pumps both amplifiers. The pump laser produces 750-mJ, 7-ns pulses at 532 nm. Image relay optics eliminate the diffraction rings and spatial inhomogeneities that develop with the free propagation of the pump beam from the Nd:YAG laser and provide a smooth pump energy distribution for both amplifiers. The protection circuit that we employ to prevent amplifying seed pulses that contain a satellite pulse reduces the repetition rate of the amplifiers from 10 to ~6 Hz.

The regenerative amplifier cavity forms a TEM_{00} waist diameter of 1.8 mm and has a round-trip time of 12 ns. Fifty-five millijoules of 532-nm energy

0146-9592/91/131001-03$5.00/0

Observation of Electromagnetically Induced Transparency in Collisionally Broadened Lead Vapor

J. E. Field, K. H. Hahn, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(Received 19 July 1991; revised manuscript received 14 October 1991)

We report the observation of electromagnetically induced transparency on the collisionally broadened resonance line of Pb vapor. By applying a 1064-nm laser beam, the transmission at 283 nm is increased by at least a factor of exp(10), with nearly all of the Pb atoms remaining in the ground state.

PACS numbers: 42.50.Qg, 32.80.Dz, 42.50.Hz, 42.65.Ky

In a recent experiment, Boller, Imamoğlu, and Harris [1] have shown how, by applying an additional electromagnetic field, an optically thick transition whose upper level decays by autoionization can be rendered transparent to a probe frequency. Such transparency is a special case of the formalism and examples given by Lambropoulos [2,3], Eberly [4,5], Agarwal [6,7], Knight [8], and their co-workers. It is also of the same nature as that observed by Whitley and Stroud [9] and Alzetta *et al.* [10] in the Rayleigh scattering of radiatively broadened systems, and is also related to the Holtsmark profile of ion broadened lines [11]. The transparency, in essence, results from the combined ac-Stark splitting and quantum interference of the dressed states which are created by the additional electromagnetic field.

In this Letter, we extend the observation of electromagnetically induced transparency to the collisionally (pressure) broadened regime. We study the transmission of the 283-nm resonance transition of neutral lead vapor with, and without, a coupling laser at 1064 nm. The energy-level diagram of lead is shown in Fig. 1. In these experiments, the self-collisional broadening of the resonance transition exceeds its lifetime broadening by a factor of 40. We observe an increase in the transmission of optically thick Pb vapor from well below our dynamic range of $\exp(-14)$ to $\exp(-4)$.

Noting Fig. 1, in the absence of the 1064-nm coupling laser, a 283-nm probe will be absorbed by the strongly pressure broadened $|1\rangle$-$|3\rangle$ transition. With the 1064-nm coupling laser present, one expects both single-photon absorption on $|1\rangle$-$|3\rangle$, and also two-photon absorption on the $|1\rangle$-$|2\rangle$ transition. This expectation is correct when the Rabi frequency of the coupling laser is much less than the square root of the product of the Doppler width of the $|1\rangle$-$|2\rangle$ transition and the detuning of the probe laser from the $|1\rangle$-$|3\rangle$ transition (6 cm^{-1}, in this experiment). When the Rabi frequency of the coupling laser substantially exceeds this product the behavior of the system is very different [1,12]. The coupling laser dresses the system so as to create the equivalent energy-level diagram shown in the inset of Fig. 1. There is now a quantum interference between the $|1\rangle$-$|2d\rangle$ and $|1\rangle$-$|3d\rangle$ absorption paths. If a collision is fast (impact regime), it establishes a coherence between states $|2d\rangle$ and $|3d\rangle$. If a perturber interacts dominantly with bare state $|3\rangle$, this coherence results in a destructive interference and, in principle, zero absorption at line center. If a perturber interacts dominantly with bare state $|2\rangle$, then the interference is constructive and the probe laser sees the Lorentzian wings of the dressed states. Equivalently, when viewed in the bare-state basis, the superposition state of the atom has negligible amplitude in bare state $|3\rangle$ and is therefore nearly unaffected by $|1\rangle$-$|3\rangle$ collisions. For a system to show electromagnetically induced transparency in the collisionally broadened regime, it is therefore essential that a perturber interact dominantly with only bare state $|3\rangle$. In the present experiment, the principle perturbers are ground-state Pb atoms. State $|3\rangle$ lead atoms interact resonantly ($1/R^3$) with these atoms with a self-broadening coefficient of 0.018 cm^{-1} per 10^{17} atoms/cm^3. State $|2\rangle$ lead atoms interact nonresonantly ($1/R^6$) with these atoms, with a broadening of 0.0012 cm^{-1} per 10^{17} atoms/cm^3.

The characteristics of the coupling laser are critical to an experiment of this type. The present experiment was chosen because of the fortunate near coincidence of the $6s^26p7s\,^3P_1^0$-$6s^26p7p\,^3D_1$ Pb transition (calculated $gf=0.98$) [13], with the 1064-nm line of the Nd-doped yttrium-aluminum-garnet (Nd:YAlG) laser. This laser (Quanta-Ray DCR-2A) was injection seeded by a diode-

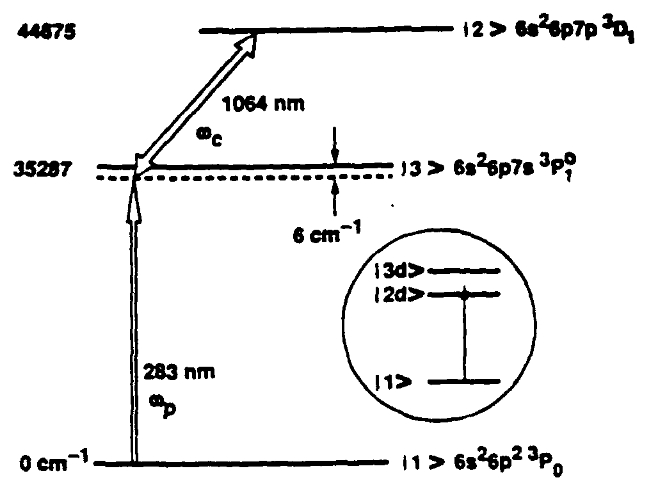

FIG. 1. Energy-level diagram of neutral Pb. Inset: Dressed-state picture.